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(54) Title: ROD-SHAPED INSERTS IN REACTOR TUBES
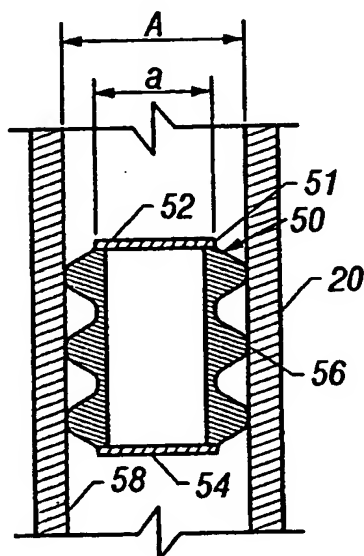
(57) Abstract: A process for catalytically reacting a fluid reactant mixture in a multitubular reactor system to form a fluid product, whereby the reactant mixture is led through at least one reactor tube (20) containing a fixed bed of solid particulate catalyst and surrounded by a heat-exchange fluid, and whereby an upstream portion of the reactor tube is dedicated to pre-heating the reactant mixture and/or a downstream portion of the reactor tube is dedicated to post-cooling the product, characterised in that the upstream portion and/or downstream portion of the reactor tube (20) contains a heat-exchanging essentially rod-shaped insert (50), the insert having a length of from 1 to 20%, preferably 1 to 50%, of the total length of the reactor tube.

WO 02/26370 A1

ROD-SHAPED INSERTS IN REACTOR TUBES

Field of the invention

The present invention relates to a process and associated apparatus and systems for catalytically reacting a fluid reactant mixture in a multi-tubular
5   reactor system.

Background of the invention

Catalytically enhanced reaction processes are used in the synthesis of hydrocarbons and their oxygenated derivatives by the catalytic hydrogenation of carbon
10   monoxide (the Fischer-Tropsch process) and in the epoxidation of alkenes. Such reactions are typically highly exothermic. They are generally performed in a vertical shell-and-tube exchanger type reactor comprising a multitude of reaction tubes, each containing a solid
15   particulate catalyst and surrounded by a heat-exchange fluid.

Typically, a production of ethylene oxide can be performed by the catalytic oxidation in the vapour phase of ethylene with a molecular oxygen-containing gas.
20   Shell-and-tube reactors used for ethylene oxidation contain several thousands of reaction tubes, each 6-15 m long and each having an inside diameter of between 20-50 mm. The ethylene oxidation solid particulate catalysts are generally based on silver supported on an inert
25   carrier material, to which promoters and co-promoters may be added. Supported ethylene oxidation catalysts comprising silver and one or more of the alkali metals K, Rb and Cs are disclosed in US-A 3,962,136 and US-A 4,010,115. Supported ethylene oxidation catalysts
30   comprising silver, rhenium, at least one further metal and optionally a rhenium co-promoter are disclosed in

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EP-B 0 266 015. A heat-exchange fluid can be a
hydrocarbon or a mixture of hydrocarbons, or it can be
water.

The reaction of ethylene oxidation is conducted at a
temperature which is broadly in the range of from 150 to
350 °C. Depending on the reactor design, the catalyst
composition, the feed composition and the further
reaction conditions a narrow reaction temperature range
has to be maintained within any given reactor. The
operation involves the pre-heating of the incoming feed
gases in the upstream portion of the reaction tubes to
the required reaction temperature, removing excess heat
during the reaction, and cooling down the effluent of the
reaction.

For greater efficiency of the pre-reaction heating
and post-reaction cooling, the upstream and/or downstream
portions of the ethylene oxidation reactor tubes are
traditionally packed with particulate material (also
termed packing). Such particulate material may or may
not be the same as the particulate catalyst used. In the
former case, expensive catalyst is being used as an
inert. In US-A 5,292,904 the use is disclosed of inert
particles as packing material in an upstream preheating
zone and in a downstream cooling zone of multi-tubular
reactors for the production of ethylene oxide. US-A
4,061,659 is directed to a process for the production of
ethylene oxide in a multi-tubular reactor, whereby the
downstream portion of the reactor tubes is filled with an
inert refractory particulate material having a surface
area of 0.1 m^2/g or less.

Japanese patent publication JP2000-169463-A envisages
the use of stainless steel packing as inert material, the
main advantage being that stainless steel is less prone
to by-product formation. It should be noted that it is
well known, *see e.g.* Wakao & Kaguei, Heat transfer in

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packed beds; Gordon & Breach, 1982), that for tubes
filled with particulates, the thermal conductivity of the
packing material has little influence on the heat
transfer under turbulent flow conditions. Therefore,
5   there would be no significant improvement of heat
transfer expected when applying (stainless) steel as
compared to low conductivity materials such as silica.

When inert particles are used in the upstream and/or
downstream portion of the reactor tubes, they have the
10   advantages of being less expensive and having a longer
useful life than catalyst particles. On the other hand,
their separation from exhausted catalyst introduces a
complication.

There exists therefore a need for the use of a
15   different kind of inserts in the upstream and/or
downstream portion of the reactor tubes. These inserts
should be at least as efficient as the known particulate
filling materials in promoting the pre-heating and/or
post-cooling of the feed gases. In particular, their
20   heat transfer coefficient should be high and the gas
pressure drop caused by them should be low. Moreover,
they should be inexpensive and easily separable from the
catalyst particles.

The use of coherent inserts of varying and more or
25   less complicated shapes is known in the art of heat
exchange, but not in chemical reactors. They are
generally designed to cause maximal turbulence of the
fluid - mostly liquid - flowing through. Indeed, they
are frequently called turbulators or turbulence inserts.
30   An example of an insert having a complicated shape is
to be found in EP-B 0 061 154, wherein an insert for
placement in a vessel is disclosed, comprising an
elongated core having a plurality of loops disposed
longitudinally therealong and angularly thereabout
35   wherein a portion of each loop lies in close proximity to

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a conceptual enveloping surface, so that laterally
rectilinear lines (L) extending from the core to the
midpoint of said portion are disposed at different
longitudinal positions along the core, characterised in

5   that each line (L) of each loop lies at an acute angle to
the core. Preferably, the core comprises two elongated
elements twisted together, a part of each of said loops
being held between the twisted together elements.

In US-A 5,454,429 a forced-flow heat exchanger is

10   disclosed, comprising a housing as part of an outer
thermal cycle and at least one tube attached in parallel
to the longitudinal housing axis as part of an inner
thermal cycle, wherein each of said tubes comprises a
loosely inserted flexible rod which is freely movable and

15   rotable in the axis and radial tube directions. The
flexibility of the rods and their being freely movable
are stated to be very essential features, since the
resulting vibrations promote a turbulent flow inside the
tubes. The rods according to this disclosure are

20   relatively thin, the preferred ratio between the inside
tube diameter and the rod diameter being from 1.4 to 2.5.
The rods occupy the entire length of the tubes.

This and the further art searched contains no
reference to the use of any type of coherent (as opposed

25   to particulate) insert for promoting the pre-heating of
reactants in a chemical multi-tubular reactor, in
particular when the reactants are gaseous and more in
particular when they comprise a carbon monoxide/hydrogen
or an ethylene/oxygen reaction mixture.

30   Summary of the invention

It has now been found that for the intended use of
promoting the pre-heating and/or post-cooling of the
reaction gases in the upstream and/or downstream portion
of multi-tubular reactors which further contain solid

35   catalyst particles, an essentially rod-shaped insert made

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of an inert material and having a length of from 1 to 20%, preferably 1 to 5%, of that of the reactor tube is at least as effective as is a multitude of packing particles occupying the same length, while having the

5   additional advantages of being inexpensive and easy to insert and to remove. Where greater heating/cooling efficiency is provided by the use of such inserts to promote pre-heating and/or post-cooling, this has the further advantage that it enables, if so desired, the

10   shortening of the heating and/or cooling zone in a catalytic reactor tube.

       The present invention therefore involves a process for catalytically reacting a fluid reactant mixture in a multi-tubular reactor system to form a fluid product,

15   whereby the reactant mixture is led through at least one reactor tube containing a fixed bed of solid particulate catalyst and surrounded by a heat-exchange fluid, and whereby an upstream portion of the reactor tube is dedicated to pre-heating the reactant mixture and/or a

20   downstream portion of the reactor tube is dedicated to post-cooling the product, characterised in that said upstream portion and/or downstream portion of each reactor tube contain a heat-exchanging essentially rod-shaped insert, the insert having a length of from 1 to

25   20%, preferably 1 to 5%, of the total length of the reactor tube.

       In particular, the reactant mixture comprises carbon monoxide and hydrogen and the product comprises a hydrocarbon and/or an oxygenate thereof, or the reactant

30   mixture comprises an alkylene and oxygen and the product comprises an alkylene oxide. More in particular, the reactant mixture comprises ethylene and oxygen and the product comprises ethylene oxide.

Brief description of the drawings

A more particular description of the invention, briefly summarized above, may be had by reference to the embodiments thereof which are illustrated in the appended drawings and described herein. It is to be noted, however, that the appended drawings illustrate only some embodiments of the invention and are therefore not to be considered limiting of its scope for the invention may admit to other equally effective embodiments.

Figure 1 is an exemplary cross sectional schematic view of a reactor having inserts disposed therein.

Figure 2 is a longitudinal cross sectional schematic view of one exemplary insert.

Figure 3 is a longitudinal cross sectional schematic view of another embodiment of an insert.

Figure 4 is a longitudinal cross sectional schematic view of another embodiment of the insert.

Figure 5 is an exemplary end view of an insert disposed in a reactor tube.

Figure 6 is a longitudinal cross sectional schematic view of one or more expandable protrusions of an insert in a retracted position.

Figure 7 is a longitudinal cross sectional schematic view of the expandable protrusions in an expanded position.

Figure 8 is a cross sectional schematic end view of the insert and reactor tube of Figure 6.

Figure 9 is a longitudinal cross sectional schematic view of another embodiment of the insert.

Figure 10 is a cross sectional schematic end view of the insert.

Figure 11 is a schematic cross sectional end view of an insert.

Figure 12 is a schematic cross sectional end view of an insert.

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Figure 13 is a graph showing exemplary results.

Detailed description of the invention

Figure 1 is an exemplary cross sectional schematic
view of a reactor having inserts disposed therein. A
5   reactor 10 comprises a shell 12 housing one or more
reactor tubes 20. A first tube plate 14 is disposed
concentrically within the shell and is coupled to the
tubes 20 to provide lateral support for the tubes.
Similarly, a second tube plate 16 is coupled to another
10   portion of the tubes 20 to provide lateral support
therefor. One or more inlets, such as inlets 22, 24, are
formed in the reactor 10 to allow reactants to enter the
reactor. One or more outlets, such as outlet 26, are
formed in the reactor 10 to allow a resulting product to
15   exit the reactor. Temperature control inlet 28 and
outlet 30 allow cooling/heating fluid to enter and exit,
the reactor 10, respectively.

The reactor 10 can be divided into a heating zone 32,
a reaction zone 34, and a cooling zone 36. The order,
20   size, and existence of the zones and inlets/outlets can
vary depending on whether the reaction is endothermic or
exothermic, the speed of the reaction, flow rate, and
other factors. A catalyst is generally disposed within
the reactor tubes 20 in the reaction zone 34.

25   One or more, and generally all, of the reactor tubes
20 have one or more inserts 38 disposed therein. While
it is feasible for a mixture of inserts to be used for a
single reactor, for economical and practical reasons, it
may be desired that all inserts will be the same in a
30   single reactor.

In some embodiments, such as shown in Figure 1, one
set of inserts 38 can be disposed in an upstream portion
of the reactor tubes 20 and another set of inserts 40 can
be disposed in a downstream portion of the reactor tubes.

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Further, it is possible for certain tubular reactors, such as those utilised in the catalytic conversion of ethylene and oxygen to ethylene oxide, that an efficient usage of the inserts proposed by the present invention is

5   to have one size of insert in the upstream portion of a reactor tube 20 and a different size of insert in the downstream portion. In such situations, the upstream insert 38 would suitably be of shorter length than the downstream insert 40, and very suitably it would be

10   approximately half the length of the downstream insert. For example, the upstream insert 38 may have a length of from 1 to 10%, or from 1 to 5%, of the total length of the reactor tube, and the downstream insert 40 may have a length of from 2 to 20%, or from 2 to 10%.

15   An inside heat transfer coefficient of the inserts according to the present invention is suitably from 800 to 4000 $Wm^{-2}K^{-}$ (Watt per m^2 per degree Kelvin), based on inner tube surface area, when measured at Reynolds numbers in the range of from 10,000 to 50,000, based on

20   void tube dimensions and superficial velocity.

The pressure drop along a portion of the reactor tube 20 occupied by the inserts 38, 40 according to the present invention is suitably from 50 to 20,000 Pa per metre, also when measured at Reynolds numbers in the

25   range of from 10,000 to 50,000, based on void tube dimensions and superficial velocity.

Figure 2 is a longitudinal cross sectional schematic view of one exemplary insert. An insert 50 can be disposed into a reaction tube 20. The insert according

30   to the present invention can have various shapes. The insert 50 shown in Figure 2 can have a core 51 with a substantially cylindrical shape. The insert 50 is preferably hollow, and closed on at least one end 52, more preferably at both ends, 52, 54.

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A cross-sectional external dimension "a" of the core
51 defines a cross-sectional external surface area of the
core 51. A cross-sectional dimension "A" of an inside
surface 58 of the reaction tube defines a cross-sectional

5   internal surface area of the reactor tube portion in
which the insert 50 is disposed. Generally, the cross-
sectional external surface area of the core 51 can be
from about 20 to 90% of the cross-sectional internal
surface area of the reactor tube 20.

10   The insert 50 can comprise surface protrusions 56,
such as rings, bars, helices, other shaped protrusions,
or a combination thereof, extending circumferentially or
longitudinally along the length of the insert. The
protrusions generally contact the inside surface 58 of

15   the reactor tube 20 and secure the insert in position
inside the reactor tube.

Figure 3 is a longitudinal cross sectional schematic
view of another embodiment of an insert. Similar to
Figure 2, an insert 50 with surface protrusions 56 can be

20   disposed within a reactor tube 20. The insert 50 can
have a conical shape. The cone can also be preferably
hollow, and closed at one end 54. The insert 50 can have
an upstream taper 53. The insert can also have a
downstream taper (not shown).

25   It is to be understood that the insert shape can vary
and can be any geometric shape as is appropriate to the
purposes of the invention.

Figure 4 is a longitudinal cross sectional schematic
view of another embodiment of the insert. An insert 50

30   is shown disposed in a reactor tube 20 on an inside
surface 58 of the tube. The insert 50 can have an
elongated core 51. As one example, the core 51 can have
a cross-sectional external surface area of from about 20
to 90%, for example about 20 to 80% of the cross-

35   sectional internal surface area of the reactor tube.

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The core 51 can comprise one or more surface
protrusions 56, such as wire mesh, brush bristles,
longitudinally positioned smooth or corrugated plates,
fins or wings of any shape and direction, or a
5   combination thereof. Generally, at least some of the
protrusions contact the inside surface 58 of the reactor
tube 20 and secure the insert in position inside the
reactor tube. In some embodiments, the surface areas
and/or thicknesses of the individual protrusions 56 can
10   increase from an outer periphery 62 of the protrusion 56
towards a base 64 of the protrusion 56 and the outer
circumference of the core 51.

Figure 5 is an exemplary end view of an insert 56
disposed in a reactor tube 20. It will be appreciated
15   that in at least some embodiments, it is not necessary
for each and every protrusion to contact the reactor tube
20, but merely that sufficient contact from the one or
more protrusions secure the inset in position during use.
For example, extended protrusions 56a can contact the
20   reactor tube 20 and other protrusions 56b may not contact
the reactor tube.

Figures 6-8 are cross sectional schematic views of
another embodiment of the insert. Figure 6 is a
longitudinal cross sectional schematic view of one or
25   more expandable protrusions of an insert in a retracted
position. Figure 7 is a longitudinal cross sectional
schematic view of the expandable protrusions in an
expanded position. Figure 8 is a cross sectional
schematic end view of the insert and reactor tube of
30   Figure 6. Figures 6-8 will described concurrently with
each other.

To place an insert 50 into a reactor tube 20, it can
be useful that the associated protrusions 56 do not
contact the inside surface 58 of the reactor tube until
35   after the insert is positioned in the tube. To assist

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contact of one or more protrusions 56, shown in
Figures 1-5, against the inside surface 58 of a reactor
tube 20 once in place, a very useful embodiment is one
wherein the insert 50 can be adjusted once in position to
5   force the protrusions against or towards the inside
surface of the reactor tube. A number of methods and
systems can be used for expanding the protrusions can be
used, including, but not limited to threaded engagements,
slip engagements, cams, beveled surfaces, gas and
10   hydraulic activation, and other methods, as would be
apparent to those with ordinary skill in the art, given the
understanding provided by the description of the invention
contained herein.

For example and without limitation, the insert 50 can
15   include a translatable member 70 that can have a twist
point 72 at one end. The twist point 72 can engage a
beveled surface 74 on the protrusion 56. The twist point
72 can be turned, once the insert 50 is in position, such
that the projections 56 are expanded within the reactor
20   tube 20 and the protrusions contact the inside surface 58
or are brought into closer proximity to the inner surface
of the reactor tube. Although not shown, it is to be
understood that the insert itself or its core can also be
expanded in addition to or in lieu of the protrusion to
25   achieve a similar result, that is, expansion of at least
some member of the insert toward the reactor tube 20.

Figure 9 is a longitudinal cross sectional schematic
view of another embodiment of the insert. Figure 10 is a
cross sectional schematic end view of the insert and
30   reactor tube of Figure 9 and will be described
concurrently with Figure 9.

An insert 56 can be disposed in a reactor tube 20.
The insert 56 can have a rod-shaped, but core-less, form,
such as a quantity of wire mesh. The insert 56 can be
35   sized to fit with its periphery adjacent to the tube's

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inside surface and so fit snugly into the reactor tube.
For example, the wire mesh can include longitudinal
members 56 and lateral members 82. The insert 56 can be
formed into an appropriate shape and size to fit within
5   the reactor tube 20.

Regardless of the particular shape and configuration,
preferably each insert is made of a metal or an alloy
which is sufficiently conductive to heat and resistant to
corrosion under the reaction conditions, such as carbon
10   steel or stainless steel. The metal/alloy of choice may
vary depending upon the reaction carried out in the
tubular reactor.

Exemplary process for use with insert

Exemplary to the operation of a process according to
15   the present invention is the epoxidation of ethylene.
The processes for the catalytic production of ethylene
oxide in the vapour phase from ethylene and molecular
oxygen are broadly divided according to the source of
oxygen used, into those using pure oxygen and those using
20   air - but the differences with regard to heat transfer
requirements are not fundamental and the present
invention can be applied in both cases.

Whether pure oxygen or air is used in the oxidation
of ethylene, the reaction gas mixture in both cases
25   comprises, besides ethylene and oxygen, an excess of
diluents such as carbon dioxide, nitrogen, argon, methane
and ethane, and a small quantity of a halide reaction
moderator such as ethyl chloride, vinyl chloride or
dichloroethane. For example, the reaction gas may contain
30   by volume 1-40% of ethylene, 3-12% of oxygen, 0-3% of
ethane, 0.3-50 ppm chlorohydrocarbon moderator and
balance argon and/or methane.

The inlet reaction gas pressure is in the range of
from atmospheric to 4000 kPa, preferably from 1000 to
35   3000 kPa. It is important that the insert according to

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the invention does not cause a substantial pressure drop over the length of reactor tube which they occupy. Preferably the pressure drop is less than about 2% of the inlet pressure.

The reaction (catalyst) temperature is in the range of from 150 to 350 °C, preferably from 220 to 300 °C. The Volume Hourly Space Velocity (VHSV) of the reaction gas mixture is in the range of from 1000 to 10000 h^{-1} (m^3 per m^3.h of packed catalyst) and preferably from 2000 to 8000 h^{-1} (m^3 per m^3.h of packed catalyst), measured at standard temperature and pressure conditions. VHSV is typically expressed in units of $hour^{-1}$, cubic meter per cubic meter-hour, cubic meter of gas per cubic meter of reactor volume (eg packed catalyst volume)-hour, or other like volume per volume-time units. The O_2 conversion level is 10-60% and the EO production (work rate) 30-400 kg/m^3 catalyst/hr.

When a hydrocarbon is used as the heat-exchange fluid surrounding the reactor tubes, its pressure is generally between 100 and 1500 kPa, preferably between 200 and 800 kPa, more preferably between 200 and 600 kPa. When the heat-exchange fluid is water, its pressure is between 1500 and 8000 kPa. The temperature of the heat-exchange fluid on leaving the reactor is generally between 200 and 350 °C, preferably between 220 and 300 °C.

The following non-limiting examples illustrate embodiments of the invention.

In Examples 1 and 2, two illustrative embodiments of inserts according to the present invention are given.

In Examples 3 to 5, the theoretical/calculated performance, in terms of effect on heat transfer, pressure drop and turbulence respectively, of the (gaseous) fluid flowing through a tubular reactor, was determined on the basis of:

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- inserts according to Examples 1 and 2 respectively, made of carbon steel having a specific conductivity of 45 $Wm^{-1}K^{-1}$,
- a 35 cm bed of particulate catalyst material, i.e alpha silver-impregnated alpha alumina rings (outer diameter 8.40 mm, inner diameter 2.97 mm, height 8.55 mm), and
- a void reactor tube.

Example 1 – insert

Figure 11 is a schematic cross sectional end view of an insert. The insert 50 included a construction of 32 bent fins as protrusions 56, welded individually around a central core 51 or annulus, which is a hollow cylinder, closed at one end or both ends and having an outside diameter of 25 mm. The thickness of the fin material is 1 mm and the total height of each bent fin is 8.1 mm. The outer diameter of the construction is 38.5 mm and its length 35 cm.

Example 2 – insert

Figure 12 is a schematic cross sectional end view of an insert. The insert 50 included a construction of a 1 mm stainless steel plate formed into protrusions 56, that is, the plate was corrugated with corrugation length of 5 mm and a corrugation height of 6.75 mm, bent around the core 51, similar to such in Example 1. The outer diameter of the construction is 38.5 mm and its length 35 cm.

Example 3 – heat transfer

A reactor tube is used, having an internal diameter of 38.5 mm and a length of about 35 cm, that being about 3% of the total length of a typical commercial reactor tube. The reactor tube is charged with the inserts of Examples 1 and 2 (each also having a length of 3% of the total length of a typical commercial reactor tube), with the ring-formed particles, or left void, and is heated to 250 °C. A gaseous mixture of methane (50%), ethylene

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(40%) and oxygen (10%) enters the reactor tube at a
superficial velocity of 1 ms^{-1}, 2000 kPa and 150 °C.

The heat transfer inside the tube charged with the
inserts of Examples 1 and 2 and the heat transfer inside
the void tube is determined according to the heat
transfer relations of V. Gnielinski, "Chem.-Ing..-
Techn.", 61, (1989), 160/61, assuming a state steady heat
transfer, a constant heat transfer coefficient at the
surface of the insert, a homogeneous material of the
insert, no heat production, no resistance in heat
conduction between the tube wall and the insert, and no
radial temperature gradients in the fluid. The heat
transfer inside the tube charged with ring-shaped
particulates ("packed bed"), is determined according to
P. C. Borman, et al., Chem. Engng. Commun., vol 114, pp.
17 to 47, under the same set of assumptions.

Figure 13 is a graph showing the results determined
from Example 3. The results show the calculated fluid
temperature as a function of distance from the reactor
tube entrance for a void tube, a tube with a packed bed
of catalyst rings, and tubes with the exemplary inserts
described in Examples 1 and 2. The x-axis represents a
distance in meters from the entrance of the reactor tube.
The y-axis shows the fluid temperature in degrees Celsius
to indicate the heat transfer along the length of the
reactor tube.

The lower curve 4 for the void tube has the lowest
rate of heat transfer. The temperature increase is about
10 °C through the reactor or about a 7% increase from the
starting temperature of about 150 °C. The next higher
curve 3, using a packed bed, shows the rate of heat
transfer of about 50 °C or about a 33% increase. The
next higher curve 1, using the insert constructed
according to Example 1, shows a greater heat transfer
ability by increasing the temperature by about 80 °C or

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about a 53% increase. The final curve 2, using the
insert constructed according to Example 2, shows the
largest heat transfer ability by increasing the
temperature by about 85 °C or about a 57% increase.

5   Example 4 - pressure drop

The same experimental conditions as specified in
Example 3 are maintained. The pressure drop, calculated
per metre length, is determined for the tube charged with
the inserts in Examples 1 and 2 and for the void tube

10 according to Karman-Nikuradze, see J. Nikuradze, VDI-
Vorschungsheft (1950) 361. The pressure drop for the tube
charged with the ring-formed particles is determined
according to S. Ergun, Chem.Eng.Prog., 48, 89, 1952.

The thus determined pressure drops are as follows.

15 -   For the void tube: 5 Pa per metre
-   For the ring-formed particles: 18000 Pa per metre
-   For the insert in Example 1: 870 Pa per metre
-   For the insert in Example 2: 485 Pa per metre.

Example 5 - turbulence

20 The same experimental conditions as specified in
Example 3 are maintained. The turbulence inside the
tubular reactor at a superficial fluid velocity of 1 m/s
was calculated, expressed in the Reynolds numbers as
follows.

25 -   For the void tube: 30516,
-   For the ring-formed particles: 9720,
-   For insert in Example 1: 10681,
-   For insert in Example 2: 5148.

From these results it is clear that, under the

30 condition of high fluid velocity used, the inserts
decrease rather than increase the turbulence inside the
reactor tube. In particular it is shown here that the
increased heat transfer, found in Example 3 for the two
inserts according to the invention (compared to the ring-

35 formed particles) is not due to an increased turbulence.

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<u>C L A I M S</u>

1. A process for catalytically reacting a fluid reactant mixture in a multi-tubular reactor system to form a fluid product, whereby the reactant mixture is led through at least one reactor tube containing a bed of solid particulate catalyst and surrounded by a heat-exchange fluid, and whereby an upstream portion of the reactor tube is dedicated to pre-heating the reactant mixture and/or a downstream portion of the reactor tube is dedicated to post-cooling the product, characterised in that said upstream portion and/or downstream portion of the reactor tube contains a heat-exchanging essentially rod-shaped insert, the insert having a length of from 1 to 20% of the total length of the reactor tube.

2. A process according to claim 1, characterised in that the reactant mixture comprises ethylene and oxygen and the product comprises ethylene oxide.

3. A process according to claim 1 or claim 2, characterised in that the insert has a length of from 1 to 5% of the total length of the reactor tube.

4. A process according to any one of claims 1 to 3, characterised in that the heat transfer coefficient of the insert is in the range of from 800 to 4000 $Wm^{-2}K^{-1}$, based on inner tube surface area, and the pressure drop along the tube portion occupied by the insert is in the range of from 0.05 and 20 $kPam^{-1}$ at Reynolds numbers in the range of from 10,000 to 50,000, based on void tube dimensions and superficial velocity.

5. A process according to any one of claims 1 to 4, characterised in that at least one insert has essentially the form of a cylinder having a core with a cross-

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sectional external surface area in the range of from 20
to 90% of a cross-sectional internal surface area of the
reactor tube and the insert comprises one or more surface
protrusions such as rings or bars or helices, or a

5   combination thereof with sufficient protrusions
contacting the inside surface of the reactor tube in
order to secure the insert in position inside the reactor
tube.

6.   A process according to any one of claims 1 to 4,

10   characterised in that at least one insert comprises an
elongated core having a cross-sectional external surface
area in the range of from 20 to 90% of the cross-
sectional internal surface area of the reactor tube,
which core comprises surface protrusions such as wire

15   mesh, brush bristles, longitudinally positioned smooth or
corrugated plates, and fins or wings of any shape and
direction, or a combination thereof, with sufficient
protrusions contacting the inside surface of the reactor
tube in order to secure the insert in position inside the

20   reactor tube.

7.   A process according to claim 6, characterised in that
the surface areas and/or thickness of the individual
protrusions increase towards the circumference of the
core.

25   8.   A process according to any one of claims 5 to 7,
characterised in that the insert has adjustment means by
which the outer surface can be expanded such that surface
protrusions are brought into contact with, or closer
proximity to, the inner surface of the reactor tube after

30   installation in the reactor tube.

9.   A process according to any one of claims 1 to 4,
characterised in that at least one insert has the form of
a rod-shaped core-less wire mesh which fits snugly into
the reactor tube.

10. A process according to any one of claims 1 to 9, characterised in that each insert is made of a metal or an alloy which is resistant to corrosion under the reaction conditions.
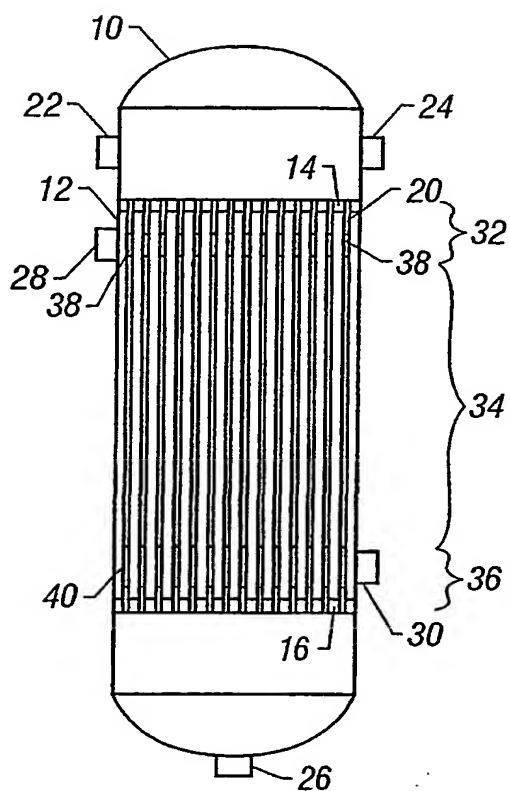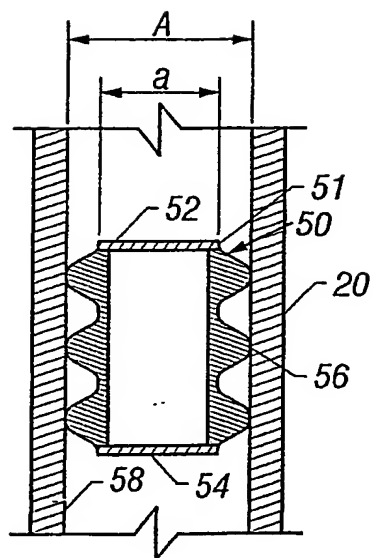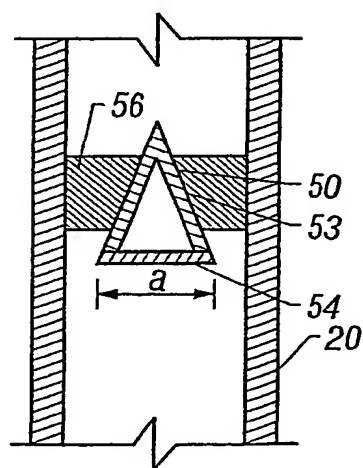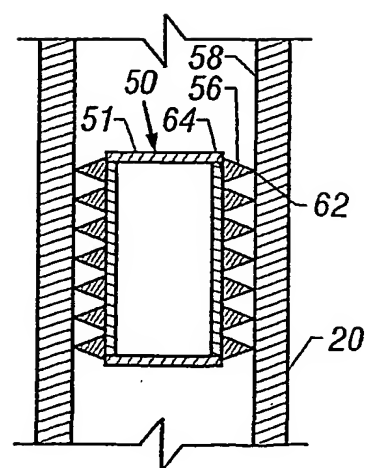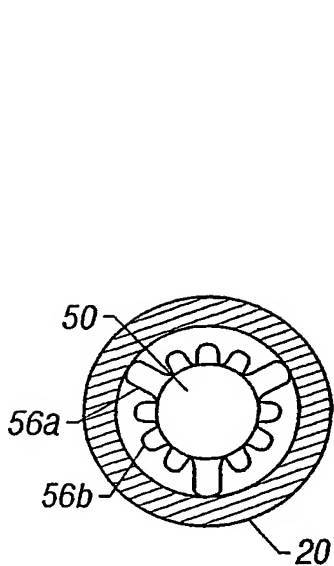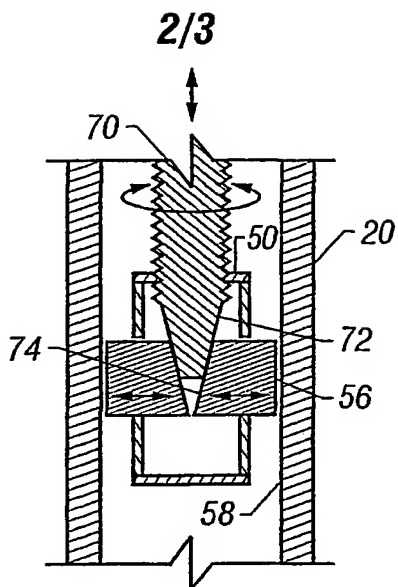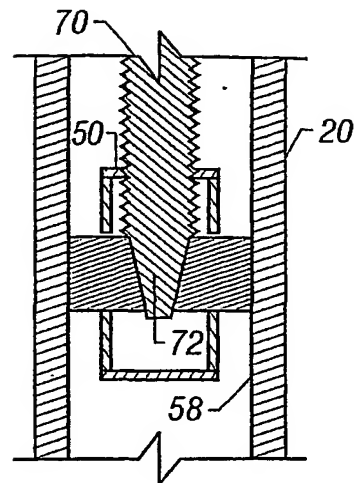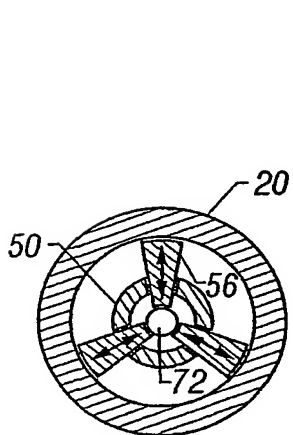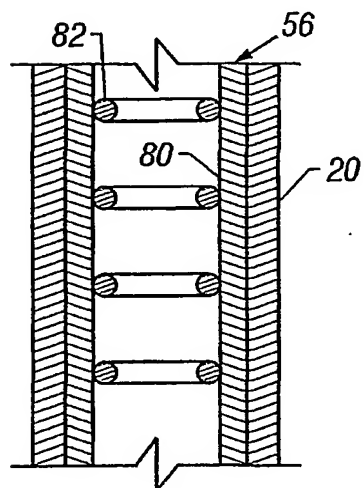
FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

FIG. 6

FIG. 7

FIG. 8

FIG. 9
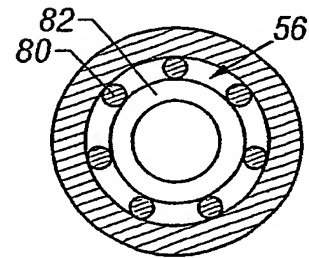
FIG. 10
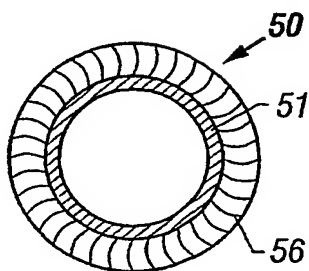
FIG. 11
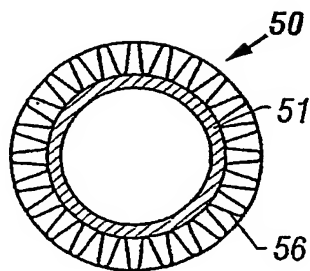
FIG. 12
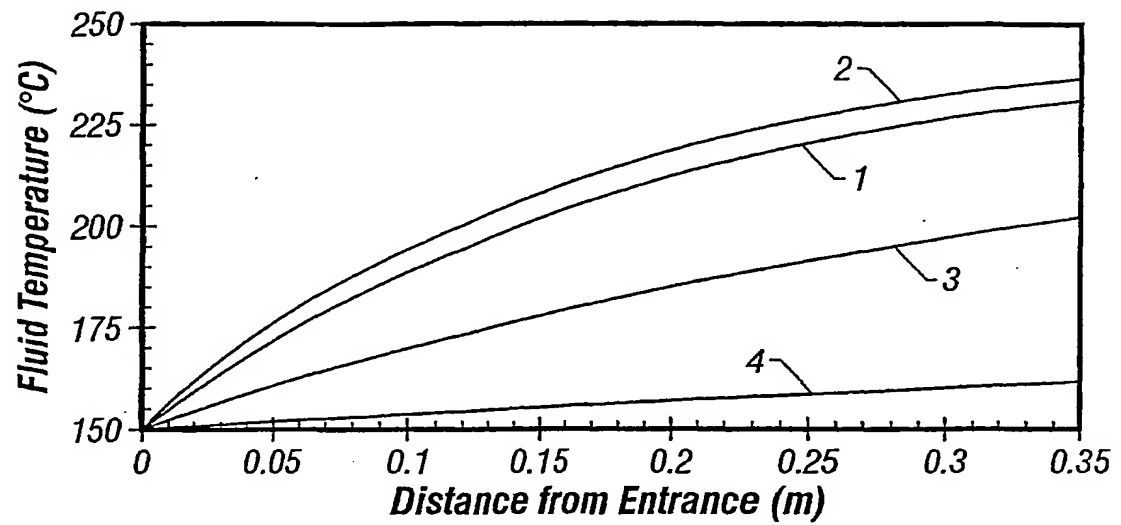
FIG. 13

INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7   B01J8/06   B01J8/00   C07D301/10   F28F1/40

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7   B01J   C07D   F28F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 061 659 A (NIELSEN ROBERT P ET AL) 6 December 1977 (1977-12-06) cited in the application column 4, line 19 -column 5, line 11 ---	1-3
A	EP 0 532 325 A (NIPPON CATALYTIC CHEM IND) 17 March 1993 (1993-03-17) page 3, linr 57 -page 4, line 18 page 5, line 8 - line 20 figure 1 ---	1,2
A	US 4 921 681 A (OZERO BRIAN J , BECKER STANLEY D) 1 May 1990 (1990-05-01) column 3, line 62 -column 4, line 34 column 6, line 29 - line 38 figure 1 ---	1-3

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[X] Further documents are listed in the continuation of box C.   [X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
5 March 2002	13/03/2002

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page 1 of 2

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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